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A Lapping Technique For Metallic, Alpha-Phase Plutonium: Achieving and Measuring Nano-Meter Roughness and Sub-Micron Flatness

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Introduction:

There are various processes and techniques by which one can achieve well polished (highly reflective) and smooth, flat (low roughness) surfaces on a wide variety of materials. Historically, precision lapping processes were first applied to optical materials for the production of lenses for telescopes and microscopes. These processes have been developed by a multitude of technicians over the centuries and reported in countless publications. These same grinding, lapping and polishing processes have been of great importance in the fabrication and structural characterization of countless optical materials and engineering structures, e.g. optics, Si wafers, precision machined surfaces, engineered devices, low-friction surfaces, mechanical systems, metallographic examination of microstructures, to name but a few.

The development of lapping and polishing processes and techniques as applied to extremely hazardous and highly (chemically) reactive materials, such as metallic plutonium (Pu), has been limited. This is due in part due to the sensitive nature of the work as applied to nuclear applications, and limited access and need to work with these materials. For the limited information regarding polishing and lapping of Pu materials that has been documented, there has been virtually no quantitative assessment of qualities such as surface roughness and flatness on the nanometer scale.

In this report we use the word "lapping" to refer to the traversing of a specimen surface across an abrasive surface in a controlled manner in order to achieve smooth/flat surfaces. We differentiate lapping from polishing, in that we define polishing as traversing of a sample surface across a non-abrasive surface that contains loose abrasive particles, e.g. diamond paste on nylon nap, in order to achieve a highly reflective surface.

In this report, we will detail a simple and repeatable lapping process as applied to alpha-phase metallic Pu. We will provide quantitative measurements of the surface roughness and flatness. We will demonstrate the ability to achieve sub-10 nm surface roughness and to lap millimeter-size areas to a flatness of approximately 1.0 μm. Additionally, we will assess the current limitations and discuss any artifacts that arise as a result of our polishing processes applied to metallic Pu materials.

Specimen Material:

The alpha-phase Pu sample material that we performed this lapping study on is of fairly standard pedigree. The alpha phase is the stable room-temperature monoclinic crystal structure phase of metallic Pu. This phase is also the highest density phase (theoretically 19.986 g/cm^3) of the five allotropic phases of solid Pu. Of the five allotropic solid phases, alpha Pu has the highest hardness value in the range of 250-500 Dph (Vickers hardness) and correspondingly, very low ductility (< 1% at room temperature). The chemical purity of our sample is ≈ 99.7%. The density of this sample material measured by a liquid immersion technique is 19.6 g/cm^3. The age of this material is ≈ 2 years.

Note: Alpha-phase Pu is notorious for being very chemically reactive to H, O and H_2O. Because of this chemical reactivity and the high health risk due to its radioactivity, handling in an inert atmosphere engineered enclosure (e.g., a glovebox with a nitrogen or argon atmosphere) is required. Furthermore, these tasks should only be performed by highly trained personnel. Thus, handling and executing processes on these materials is technically challenging.

Supplies, Equipment and Instrumentation:

The supplies and equipment for this study are commercially available, and have the added advantage that they are small in size, thus requiring limited room in a glovebox enclosure. Figure 1 is a photo of all of the supplies and equipment that are necessary in order to perform the lapping procedure. The lapping films (ref. 1) are an abrasive media where the diamond particles are mono-layer in nature and well-adhered to the surface of the plastic film; they do not come loose during the lapping process. The non-adhesive backed, 8 inch diameter, diamond lapping film grit sizes used for this study are 30, 9, 3, 1 and 0.5 μm. The lapping lubricant used is 200-proof ethanol. The gravity-feed lapping device is a Model 145 available from South Bay Technology (ref. 2). This lapping device has several important features:

1) a gravity-feed central sliding cylinder with a precise sliding fit such that minimal rocking of the sample on the surface of the lapping film is achieved, 2) ability to control the weight on the sample, 3) tungsten-carbide feet for minimum wear; these features are ground precisely perpendicular to the sliding central cylinder, 4) a removable specimen mount that is indexed to the central sliding cylinder allowing for removal of the mounted specimen for measurement and then reattachment with precise alignment. Figure 2 show the lapping device disassembled with descriptions for the parts. A micrometer with digital readout and a measurement precision of ± 1 μm is required to monitor the amount of material removal (Ref. 3). A weight of know mass for gravity feed pressure on the sample is also needed. Various size weights can be fabricated to change the pressure on the sample surface. For this study, a Cu weight was fabricated with a weight of ≈ 100 grams. An ultra-flat lapping plate is used for supporting the lapping film.

Quantitative surface roughness and flatness measurements are made using a Zygo NewView 7000 white-light interferometer (ref. 4), figure 3. The safe transfer and handling for measurement on the Zygo instrument is facilitated by encapsulating the sample in a custom made enclosure (figure 4). The enclosure contains a transparent window that the interferometer can "see" through, inset of figure 4. The interferometer is equipped with a 5x Michaelson objective, with the ability to insert a sample of the enclosure's window material in the reference leg. Thus, the interferometer compensates for the enclosure's window, and acquires high-precision data from the sample surface, with no requirement to account for the window.

(*) Additional diamond lapping film grits are available, i.e. 12, 6, 0.25 and 0.1 μm but were not included in this study.

Preliminary Checks:

1) Lapping fixture is ultrasonically cleaned and free from debris that can come loose and scratch the surface of the specimen.
2) Specimen mount is clean and free of burrs so that it will make solid contact in the lapping device.
3) Lapping films are clear of debris.
4) Glue is not beyond its expiration date and is at room temperature just prior to use.
5) Lapping plate is clean and free of nicks, burrs, and particulates.
6) Micrometer faces are clean and free of burrs and particulates that would cause poor contact with specimen surface resulting in inaccurate measurement and scratching of the specimen surface.
7) The use of a gauge block for checking the calibration of the micrometer is available. Measuring the gauge block is also a good practice in order to assess one's measuring technique.
8) Weigh sample, lapping mount, floating centerpiece and mounting screw of the lapping device, and the weight. (**note:** no need to weigh

the housing of the lapping device, it does not provided any weight on the sample during lapping).

9) Estimate or measure surface area of the specimen that will be in contact with the lapping film.

10) Be prepared to estimate the number and size of the “figure-8”patterns per minute that you will perform so that surface lapping speed can be calculated. See figure 5 for example.

The Lapping Procedure:

The following procedure is for the reproducible production of flat surfaces on metallic alpha Pu and the evaluation of surface roughness (R_a and RMS) as a function of diamond lapping film grit size. The gravity-feed lapping device will be used in a mode that we refer to as “gravity feed mode”, as opposed to a mode that we will call “drop and lock” and will be described in a second procedure.

1) Begin by thoroughly cleaning the lapping plate, front and back side of the lapping films, specimen mount, gravity-feed lapping device (GFLD), and Cu weight. **Note:** Work surfaces, lapping supplies and GFLD need to be as particulate-free as possible.
2) Manually lap the backside of the specimen mount to remove any macroscopic high points. **Note:** This will allow the mount to properly re-index to the GFLD. A uniform 30 μm diamond lapping film finish is all that is needed for this step.
3) Re-clean specimen mount.
4) Attach specimen mount to GFLD.
5) Place 30 μm diamond lapping film on lapping plate. **Note:** No liquid or adhesive is used to adhere the lapping film to the lapping plate for reasons that will be described in the discussion section.
6) Wet the central region of the lapping film with a few milliliters of ethanol.
7) Gently place GFLD onto the lapping over the wet region. **Note:** The edges of the tungsten-carbide feet on the GFLD are very sharp and can easily cut or scratch the lapping film, thus rendering it useless.
8) Place the 150 g weight on top of the GFLD.
9) Guide the GFLD in a figure-8 pattern over the surface of the lapping film. **Note:** Use very little downward hand-finger pressure to keep the GFLD in contact with the lapping film. Pressing too hard may lead to scratching or creasing of the lapping film, thus rendering it useless.
10) Continue lapping until the specimen mount is uniformly scratched across it surface. **Note:** This process will ensure that the specimen-mounting surface will be co-planar with the tungsten-carbide feet of the GFLD.
11) Remove the specimen mount and clean it.

12) Mount the specimen onto the specimen mount using Sally-Hansen cyanoacrylate brush-on glue and allow to cure. **Note:** This may take several hours, or even longer in an inert atmosphere enclosure.
13) Place a clean 30 μm diamond lapping film onto the clean surface of the lapping plate.
14) Apply several milliliters of ethanol.
15) Place GFLD, with **no** specimen mount attached, onto wet lapping film.
16) In a figure-8 pattern guide the GFLD over the central region of the lapping film for 1-2 minutes. Add ethanol as needed. Feel (with your fingers) for any bumping of the GFLD as it is guided over the surface of the lapping film. Additionally, watch for the appearance of small black dots on the surface of the lapping film. See figure 6. **Note:** These black dots are as a result of microscopic particulate between the lapping plate and lapping film. These may leave scratches in the surface of the specimen and adversely affect edge rounding. The particulate should either be removed, or avoided during lapping. This step (16) should always be performed when placing any lapping film onto the lapping plate prior to lapping of the specimen.
17) Mount specimen onto the GFLD.
18) Re-wet lapping film.
19) Place GFLD onto the lapping film and begin lapping in a figure-8 pattern in the central region of the lapping film. Use same precautions as noted in step 9 and 17. Check occasionally and continue lapping until there is a uniform scratch pattern over the entire surface of the specimen. **Note:** If the figure-8 pattern is composed of approximately 2 inch diameter circles and the circular rate is approximately 1 cycle/sec a surface speed of approximately 32 cm/sec will be achieved, which is similar to the surface speeds utilized in this study.
20) As required: A) measure the thickness of the specimen mount + specimen. B) Repeat at timed intervals and uniform lapping speed and pattern so that material removal rates can be determined. C) Measure surface area of specimen so that weight per unit area can be determined.
21) For hazardous or reactive specimens, encapsulate in a windowed enclosure and transfer to surface measuring instrumentation. (See metrology procedure)
22) Measure surface roughness.
23) Repeat steps 14 thru 22 for other diamond lapping film grit sizes. **Note:** Thoroughly clean GFKD and specimen before proceeding to next diamond grit size. For the 0.5 μm diamond-lapping film no lubricant was used. Lapping was performed dry to prevent hydride evolution on the surface of the specimen.

Metrology: (short version, see attachment A for Zygo user instructions)

1) Load and seal mounted specimen into Zygo encapsulation device. Figure 4.
2) Remove safely from glovebox antechamber and decontaminate exterior while in fumehood. Check for external alpha contamination. Label and package for transport to Zygo instrument.
3) Assure that the glass-compensation widow is inserted in the reference leg of the objective.
4) Assure that the Z-stop is set so that the objective lens will not crash into the encapsulation holder window.
5) Focus, find interference fringes, level sample, perform bi-polar scan and save data.
6) Remove encapsulation device, alpha survey external surface and stage area, re-package and return to RMA for removal of mounted sample in glovebox.

Results:

Table 1.0 contains the results of the evaluation of surface roughness (R_a and RMS) in microns (μm) for five different diamond lapping film grit sizes: 30, 9, 3, 1 and 0.5 μm. In this table it can be seen that the parameters for weight (233.5 grams), surface area (9.52 mm^2), and surface speed (32 cm/sec) were held constant; the one independent variable is the grit size (microns); and the measured outcomes are surface roughness, surface flatness (maximum change in surface height (um) over a 3 mm liner distance) and material removal rate (μm/min).

Figure 7 shows series of surface height plots depicting measured surface roughness for the five different diamond-lapping films. It can be seen that for each diamond grit size lapping film the surface has a correspondingly smaller surface roughness. Figure 8 is a series of surface images where color correlates to height at each pixel location on the surface. The decrease in the number and size of the resolvable scratches decrease with diamond grit size as well. Overlaid on these surface maps is a (diagonal) line-trace. The line trace corresponds to the line-profile plots in figure 9. Figure 9 shows pixel-by-pixel the measured height along a line-trace drawn in the surface images (line is anchored with open triangles). Again, the line-profiles clearly indicate the trend of lower roughness as a function of smaller diamond grit size lapping film. It should be noted that although the roughness and peak-to-valley numbers are slightly higher for the 0.5 μm diamond-lapping film than for 3 and 1 μm films, a close inspection of these specific line-profile plots in figure 9 indicates that the high frequency roughness appears to be lower for the 0.5 μm film. The higher values for the 0.5μm films are as a result of more round-off (or long frequency roughness) of the specimen surface.

Conclusion:

We have developed and documented a procedure for lapping low roughness and flat surfaces on metallic, alpha phase, Pu. Surface roughness of < 10 nm have been achieved. A flatness of less than 1.0 µm height change over a 3 mm diameter area has been achieved as well. We have also developed a safe way of performing white-light interferometry on these Pu specimens, thus enabling quantitative surface roughness characterization at the 1 nm scale. By using commercially available devices and supplies we were able to perform the lapping procedures within a small footprint of a glovebox enclosure, and such that it is repeatable by a skilled glovebox technician.

Figures and Tables:

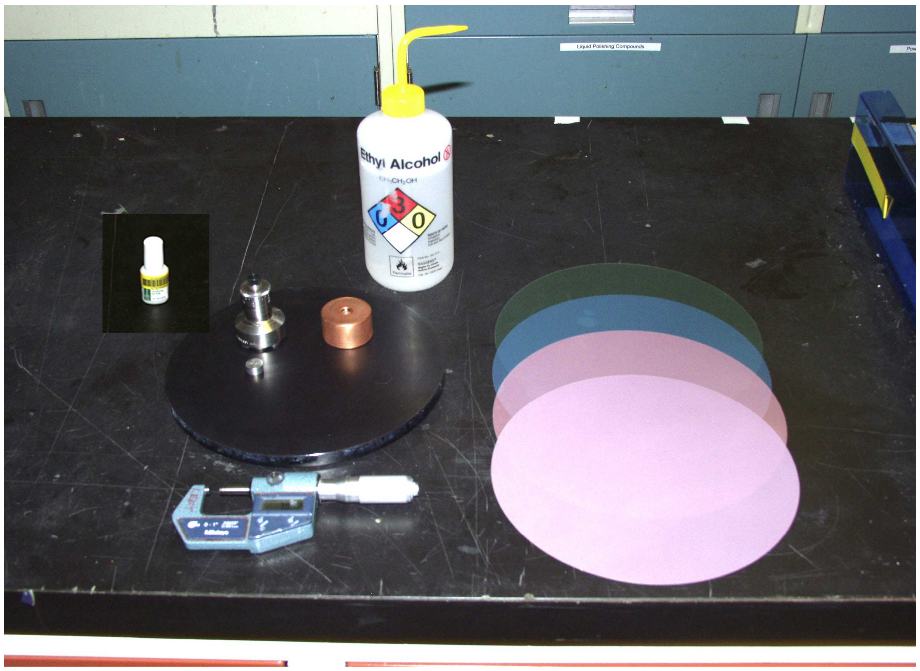

Figure 1. Supplies and instruments: micrometer, lapping films, lapping plate, lapping device, sample mount, weight, ethanol and glue. Lapping films are color coded for the diamond grit size in microns.

Figure 2. Photograph of a disassembled lapping device. Parts: (a) lapping mount showing backside with indexing hole, and center threaded hole for locking in place, (b) tungsten-carbide feet, (c) centerpiece floating rod, (d) screw rod for locking specimen mount in place.

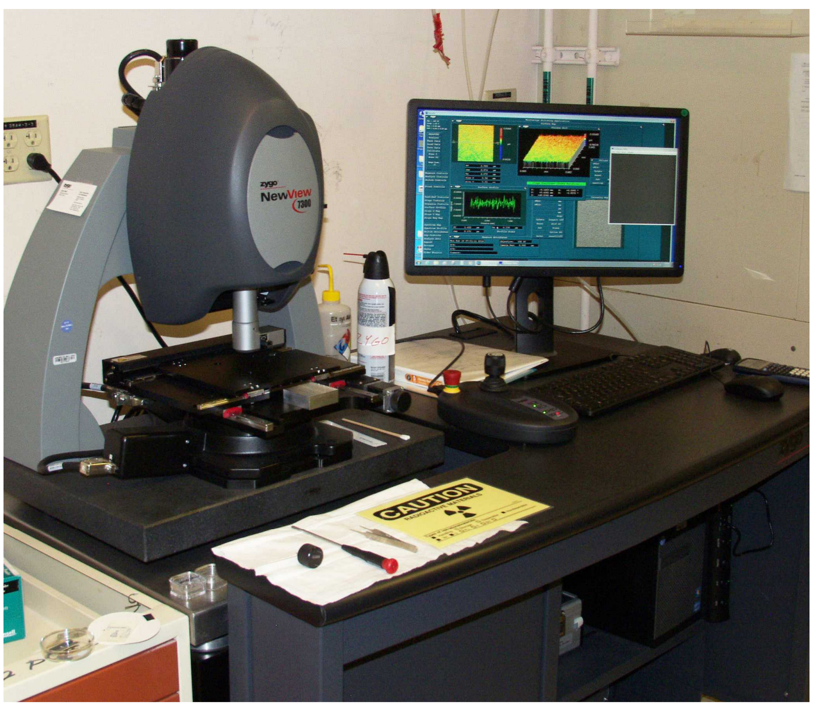

Figure 3. Zygo NewView 7000 white-light interferometer.

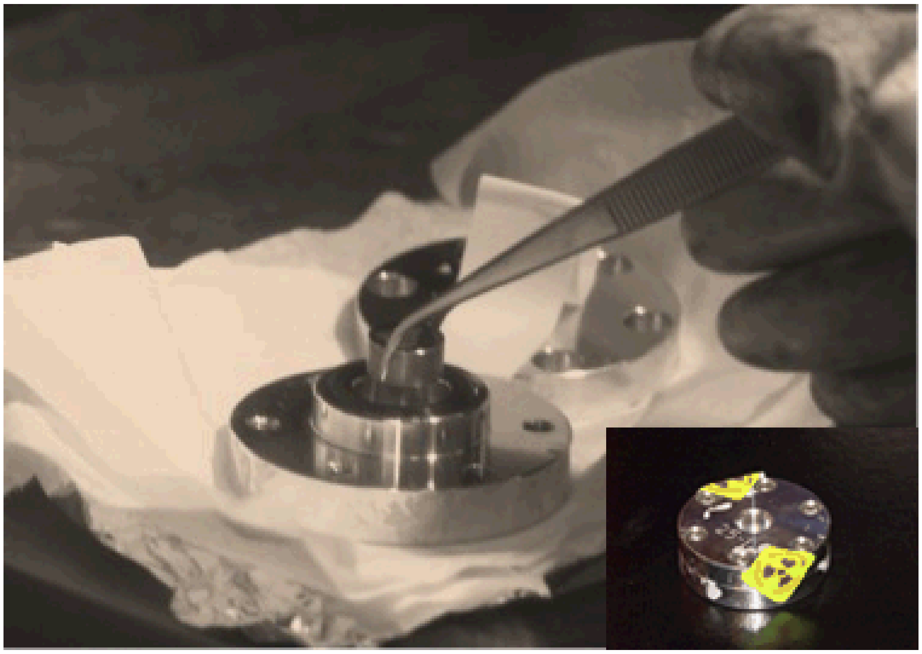

Figure 4. Image of a specimen mount (with specimen) being loaded into an opened Zygo encapsulation specimen holder in the ante-chamber of the glovebox. Inset image is of the sealed encapsulation specimen holder with top-center window.

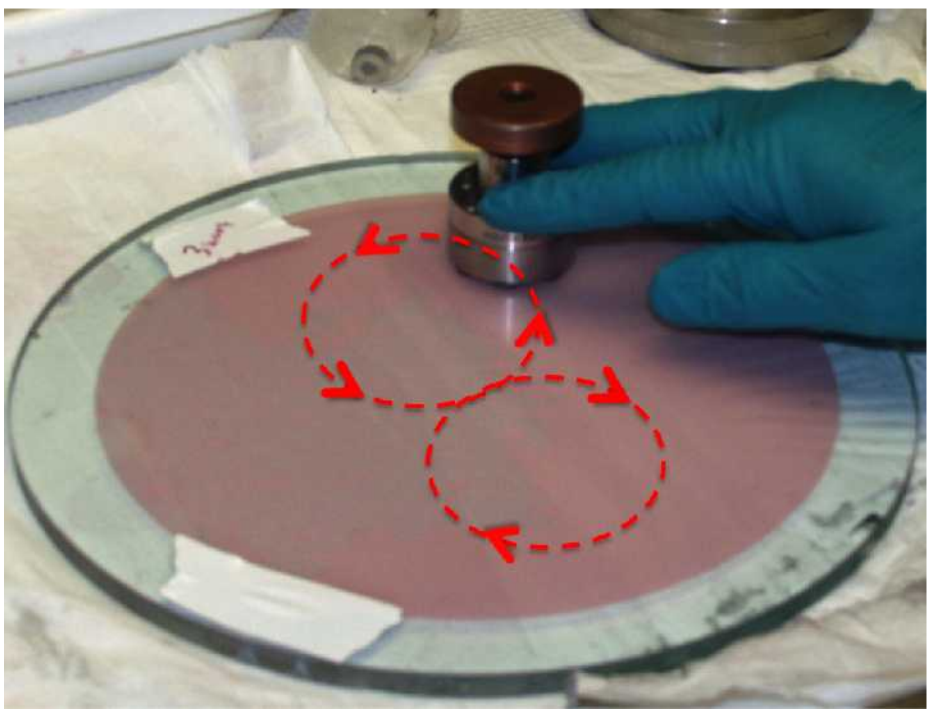

Figure 5. Hand lapping with the Model 150 gravity-feed lapping device in a figure-8 motion pattern.

Figure 6. Photograph showing black dots on surface of the lapping film as a result of microscopic particles trapped in between the lapping plate and lapping film.

Diamond lapping film grit size	Removal rate (µm/min)	Surface Roughness R_a (µm)	Surface Roughness RMS (µm)	Flatness @over a 3mm length (µm)
30	11	0.199 +/-0.010	0.266 +/-0.011	1.215
9	14	0.052 +/-0.010	0.081 +/-0.015	0.861
3	3.3	0.009 +/-0.003	0.011 +/-0.004	0.754
1	3	0.010 +/-0.007	0.012 +/-0.007	0.870
0.5	≈1	0.008 +/-0.004	0.012 +/-0.005	0.866

Table 1. Surface removal rates, surface roughness (R_a and RMS), and flatness as a function of diamond lapping film grit size.

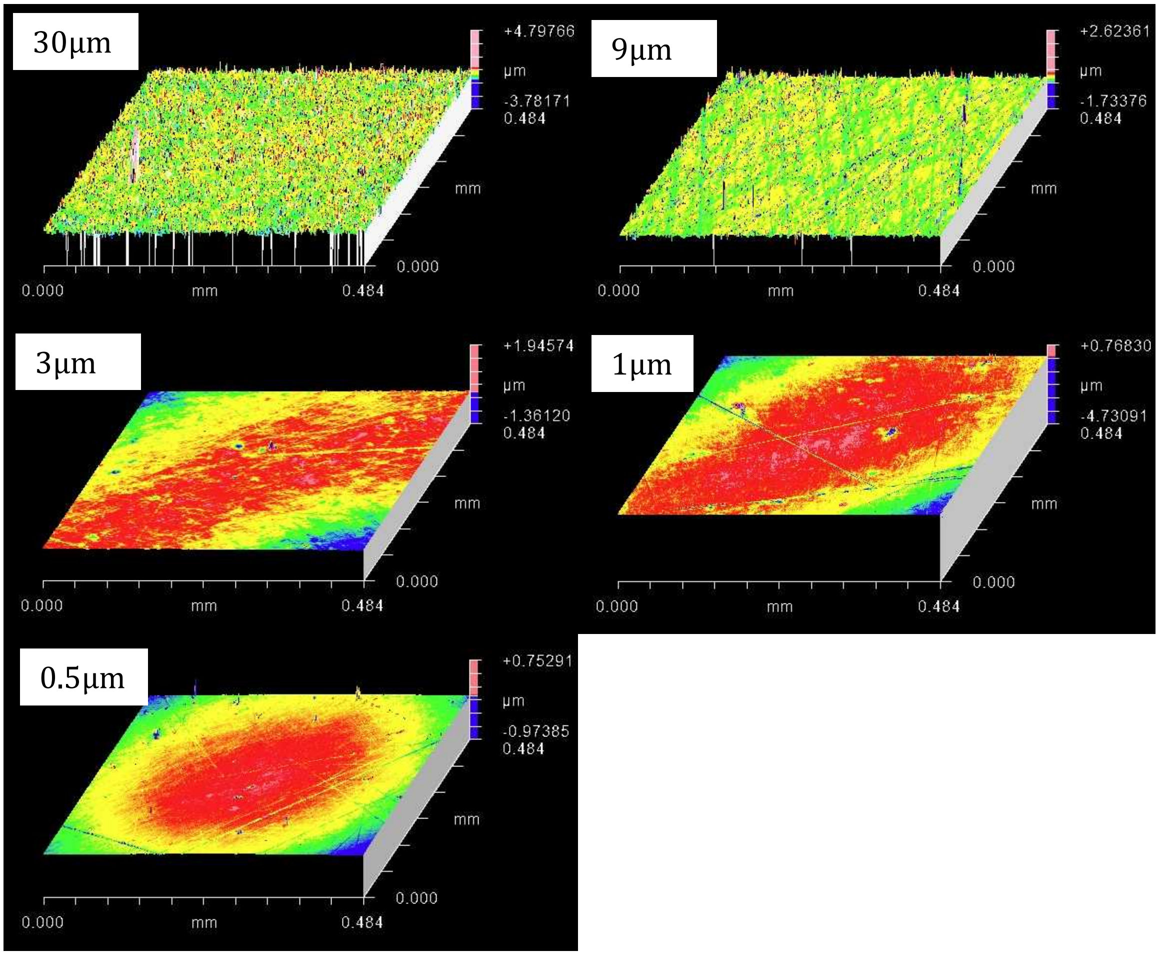

Figure 7. Surface height plots for 30, 9, 3, 1 and 0.5 µm diamond lapping films as measured on the Zygo white-light interferometer. **Note:** vertical scaling for all plots is not the same.

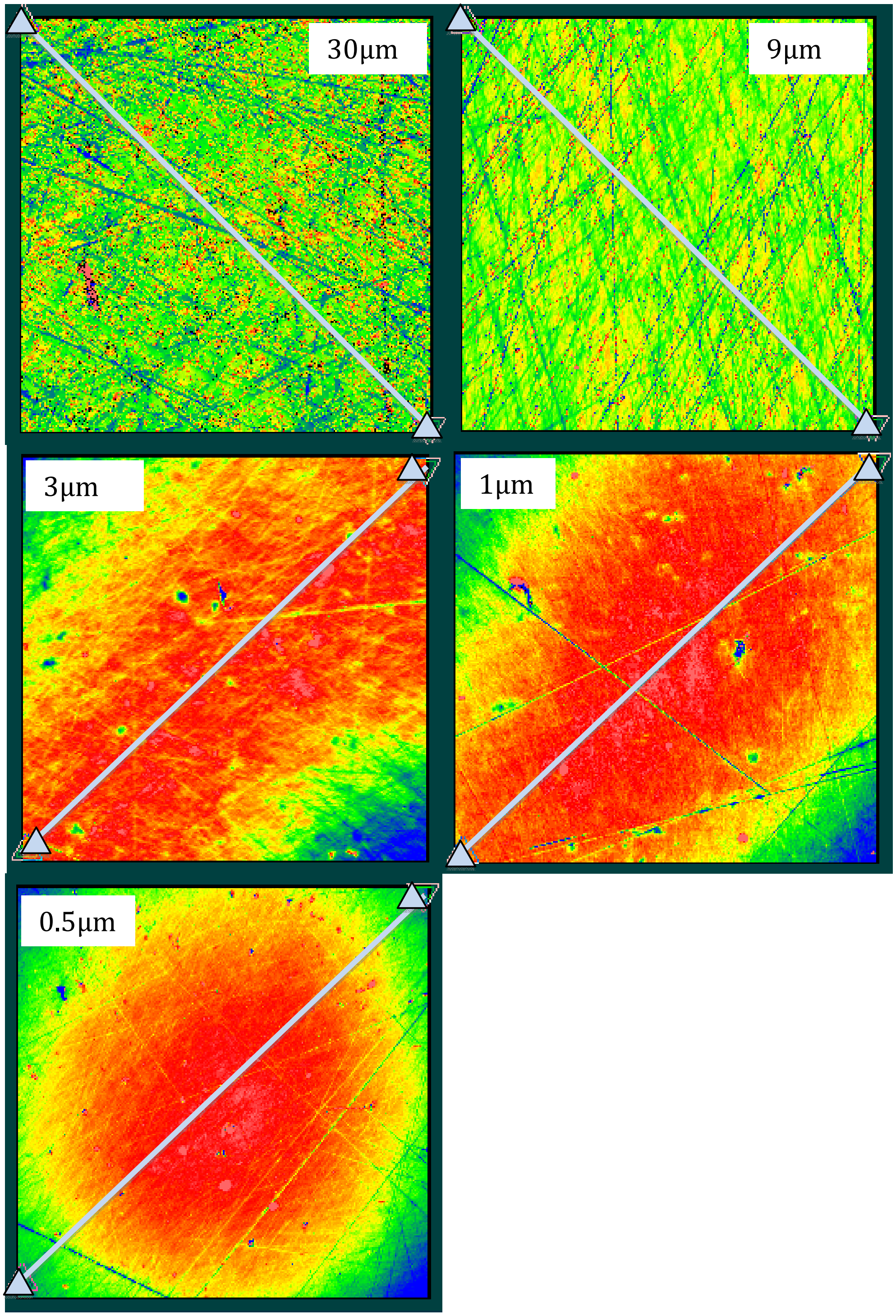

Figure 8 Surface images for 30, 9, 3, 1 and 0.5 µm diamond lapping films as measured on the Zygo white-light interferometer. Each image corresponds directly to the surface profile plots in figure 7. Diagonal line corresponds to each line profile plot in figure 9.

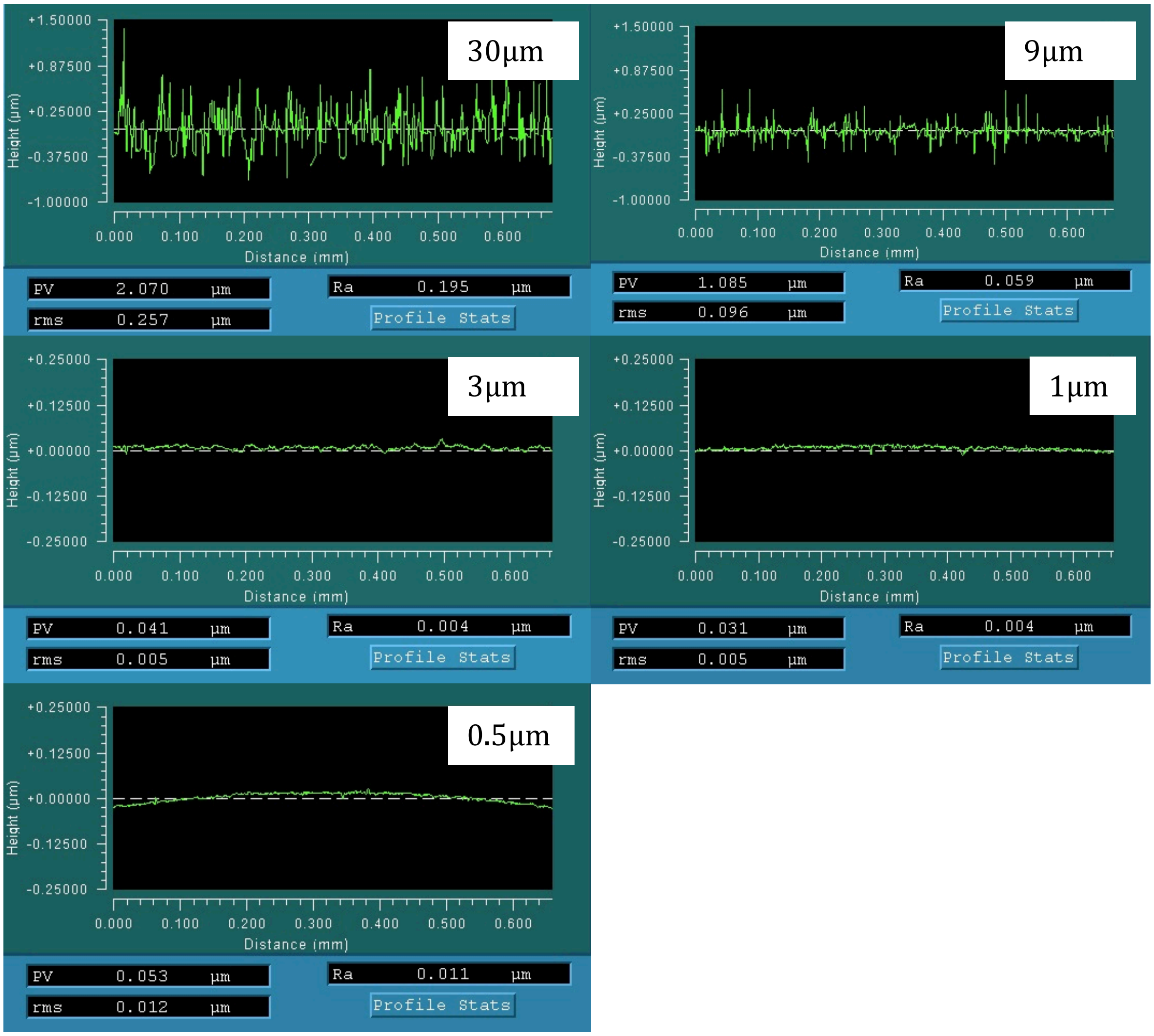

Figure 9 Line-profile plots for 30, 9, 3, 1 and 0.5 μm diamond lapping films as measured on the Zygo white-light interferometer. **Note:** not all vertical scales are similar in range.

References:

(1) Allied High Tech Inc. http://www.alliedhightech.com/
(2) South Bay Technology Inc. http://www.southbaytech.com/
(3) Mitutoyo Inc. http://www.mitutoyo.com/
(4) Zygo Inc. http://www.zygo.com/?/met/profilers/newview7000/

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Attachment A

Zygo New View 7300 general user instructions

1. Verify PC is **ON**. The computer should always be on and is not connected to the Lab Network.
2. Sample should have a reflective surface. (if not then it may need to be sputter-coated w/ Au e.g.)
3. On the **Desktop** click the **Metropro** program then the **Micro7K.app** .
   a. Two (2) windows will open.
      i. **Live Display**
         1. If the screen is **RED,** there is too much light. Press the **F5** key, to auto adjust the illumination, as necessary.
      ii. **Metropro**
         1. Check that there is illumination (**green light)** reflecting off of the surface of your sample.
4. Joystick controls
   a. Rotating the **joystick** left (CCW) or right (CW) moves the objective up and down (Z-axis) respectively.
   b. **Z-Stop** button—Sets the lowest position of the objective lens to prevent the objective lens from damage if it runs into your sample or stage surface.
   c. **X/Y** button lit—Tilting the **joystick** moves the stage; left-right (+/- X axis) or back-forward (+/- Y axis)
   d. **P/R** button — Tilting the **joystick** tilts the stage (pitch/role) for leveling.

   NOTE: Once the **Z-stop** is set an alarm will sound and the objective will not move down. To shut off the alarm, rotate the **joystick** clockwise (CW).

5. Setting the Z-Stop—Preventing damage to the objective lens.

a. Press the **Z-Stop** button, to reset the **Z-stop** location. The **red light** on the **Z-stop** button will flash.
b. Using the **joystick**, move the objective to approximately 1 cm above the sample.
c. Press the **Z-Stop** button, again, to set the **Z-stop** location. The light will change to green when the **Z-stop** is set.
d. While watching the objective lens at the specimen, move the objective lens slightly up by rotating the **joystick**, attempt to move the objective below the **Z-Stop** by rotating the **joystick**. The alarm should sound, if the alarm does not sound repeat step 5 until the **Z-stop** alarm sounds.

6. Obtaining and Focusing the sample
   a. Using the **joystick**, move the objective lens to approximately 14 mm above the sample, until the image in the **Live Display** window is ≈ in-focus.
      i. Should the image on the **Live Display** window appear **RED or have RED pixels**, press the **F5** key to auto adjust the illumination.
   b. Using the **joystick** (step 4c) translate the sample to the area of interest.
   c. Re-focus the image as needed (step 4a).
   d. On the right side of the Zygo head pull out the **F-Stop** knob.
      i. **Live Display** will have an image of the aperture.
   e. Bring the edge of the aperture into focus using the **+/-** "FOCUS" thumb wheel located on the right side of the objective lens.
      i. The aperture should have clean, sharp edges.
   f. Push the **F-Stop** knob in.
7. Locating and maximizing the contrast of the interference fringes.
   a. Using the "**THK"** thumb wheel located on the front of the objective lens, slowly adjust while observing the **Live Display** for interference fringes.
   b. Once found, now maximize contrast via iteration of the "THK" and "FOCUS" knobs. Slowly adjust these **+/-** thumb wheels to obtain the greatest contrast. **HINT:** Pick a + direction on the FOCUS knob then bring back the fringes using the – THK knob. Do this several times and notice if the fringe contrast is increasing or decreasing. If decreasing, then iteratively choose the opposite -/+ directions of the THX and FOCUS knobs until maximum contrast. Adjust illumination intensity (F5) as needed.

NOTE: If the surface of the sample is very rough and/or not very reflective, it may be difficult to locate and maximize the contrast of the interference fringes. Consider performing this step on a polished witness surface first.

NOTE: Move the **+/-** thumb wheel in the + direction, the THK thumb wheel should be moved in the – direction and vice versa.

8. Obtaining data.
    a. Click on the **Metropro** window to make it the active window.
    b. At the upper left corner of the window is a button that states "**None**", click to change to **10X GC**.
    c. Click on the Measure Controls button.
        i. Image zoom = 1X
        ii. Min. mod. = 0.1
            1. Scan Controls
                a. Bi-Polar Scan length = Should be set larger than the estimated peak to valley range of the sample surface.
    d. Click on the Analyze Controls button.
        i. Remove: This setting removes the background. Start with either "none" or "plane".
    e. Click on Measure button—Starts a scan

NOTES:

PV = Peak to Valley
RMS = Root/Mean/Square
RA = Roughness measurement
NOTE: Black pixels in the image or on the line graph = no data. Possible causes; vertical surfaces too steep, scan range too short and/or local regions not reflective.

9. To save the raw data.
    a. Click Save Report
    b. Directory to Desktop/User data
    c. Use current or create a new folder.
    d. Name the file
    e. Click Save

NOTE: Ensuing scan will over-write current data file. Save each scan individually.

10. To save an image of the data window.
    a. Right-click and hold on the **Metropro** title bar
    b. Drag to choose Print

i. Destination
    1. File
ii. Image Format
    1. .tif
iii. Background
    1. Color
iv. Print—Opens a file save window
    1. Desktop/User data/Users folder/Name the file
    2. Add the .tif extension
    3. Save